Elastic Constants of Incommensurate Solid ^{4}He

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We study the elastic properties of incommensurate solid ^{4}He in the limit of zero temperature. Specifically, we calculate the pressure dependence of the five elastic constants (C_{11}, C_{12}, C_{13}, C_{33}, and C_{44}), longitudinal and transversal speeds of sound, and the $T = 0$ Debye temperature of incommensurate and commensurate hcp ^{4}He using the diffusion Monte Carlo method. Our results show that under compression the commensurate crystal is globally stiffer than the incommensurate, however at pressures close to melting (i.e. $P \sim 25$ bars) some of the elastic constants accounting for strain deformations of the hcp basal plane (C_{12} and C_{13}) are slightly larger in the incommensurate solid. Also, we find that upon the introduction of tiny concentrations of point defects the shear modulus of ^{4}He (C_{44}) undergoes a small reduction.

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I. INTRODUCTION

An intriguing resemblance between the dependence of the shear modulus (SM) and torsional oscillator (TO) frequency changes on temperature, amplitude, and concentration of ^{3}He impurities, has been experimentally observed in solid ^{4}He at low temperatures.[1] Crystal defects are clearly involved in both phenomena, however how SM and TO fluctuations are exactly related remains yet a puzzle. Day and Beamish identified the stiffening of solid helium with decreasing temperature, i.e., increase of its shear modulus, with the pinning/unpinning of dislocations by isotopic impurities. Subsequent experiments have confirmed Day and Beamish interpretations[2] although recent elasticity measurements on ultrapure single crystals seem to suggest that SM variations cannot be uniquely understood in terms of mobile dislocations.[3,4]

Torsional oscillator anomalies were first interpreted as the mass decoupling of a certain supersolid fraction,[5,6] a counterintuitive physical phenomenon that Andreev and Lifshitz already proposed in solid helium more than 40 years ago.[7] Supporting this view is the fact that TO anomalies appear to occur only in bulk ^{4}He.[8] Nevertheless, the supersolid interpretation of TO anomalies appears to leave open its connection to SM fluctuations and diverse theoretical arguments and hypotheses have been put forward in an attempt to simultaneously rationalize the origins of both anomalies. Anderson, for example, proposes that supersolidity is an intrinsic property of bosonic crystals, which is only enhanced by disorder, and that the elastic anomaly is due to the generation of vortices at temperatures close to the supersolid transition.[9] From a diametrically opposite standpoint, Reppy has argued that the TO behavior is caused by an increase of the ^{4}He shear modulus which mimics mass decoupling by stiffening the TO setup.[10] Other scenarios somewhat more reconciling with the original TO and SM interpretations have been also proposed in which for instance mass superflow is assumed to occur in the core of dislocations only when these are static.[4,11]

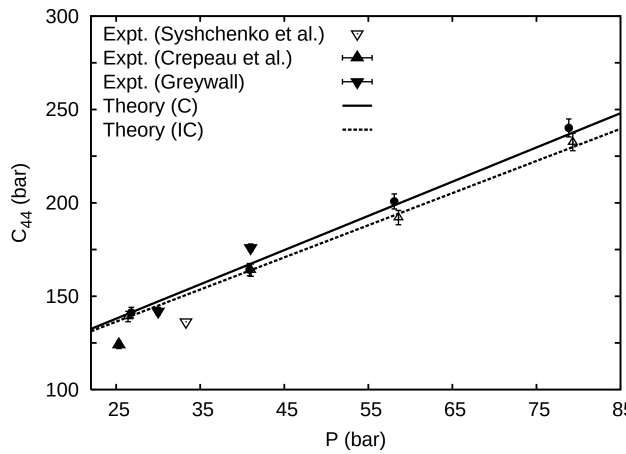


FIG. 1: Shear modulus results obtained for C and IC hcp ^{4}He expressed as a function of pressure. Experimental data from Refs. 29, 31, and 32 are shown for comparison. Solid lines represent linear fits to DMC results (see text).

As it can be appreciated, definitive conclusions on the roots of SM and TO anomalies remain contentious. In a recent paper, Chan *et al.*[12] have shown that for solid ^{4}He in vycor the nonclassical moment of inertia (NCRI) disappears if the TO setup is designed in such a way that is completely free from any shear modulus stiffening effect. This result seems to show that NCRI can be totally attributed to elastic effects and not to the existence of a supersolid fraction.[13] On the other hand, a recent experiment in which DC rotation was superposed to both TO and SM measures suggested that the cause of both anomalies below a critical temperature could have different microscopic origins.[14] Also, the source of a small peak in the specific heat of ^{4}He[15] at temperatures close to that at which TO and SM anomalies appear remains yet unexplained.

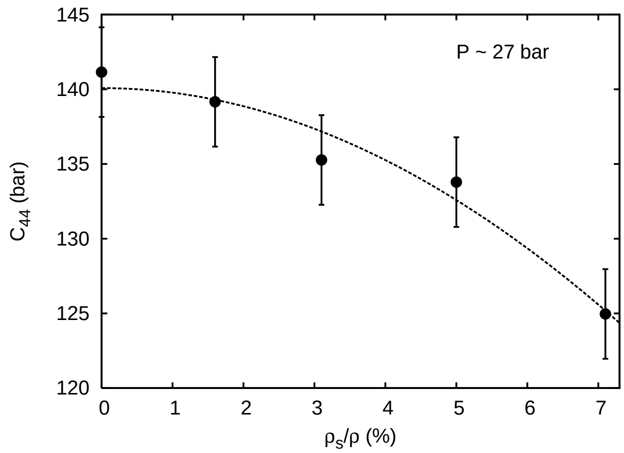


FIG. 2: The shear modulus of IC solid hcp ^{4}He expressed as a function of the superfluid fraction at pressures close to melting. The dashed line is a guide to the eyes.

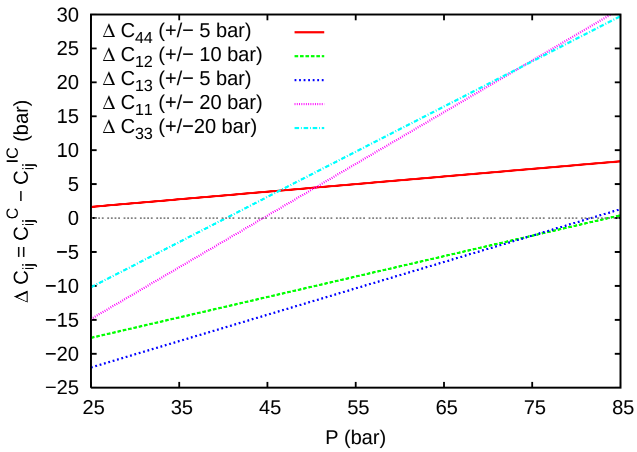


FIG. 3: Elastic constant differences between C and IC crystals ($\rho_s/\rho = 2$ %) of hcp ^{4}He expressed as a function of pressure. The size of the error bars are indicated within the parentheses. Positive ΔC_{ij} values indicate softening of the corresponding elastic constant in an hypothetical vacancy-induced normal-to-supersolid phase transition.

In this work, we study the change in the elastic constants of solid ^{4}He caused by the presence of small point defects concentrations, n_v, of $0.5 - 2.0$ % . As it has been shown, the presence of vacancies induces a finite superfluid fraction in the crystal (incommensurate crystal, IC) so that we can theoretically compare the elastic constants of a supersolid with those of the perfect crystal (commensurate crystal, C). In particular, we estimate the pressure dependence of the elastic constants C_{ij}'s (C_{11}, C_{12}, C_{13}, C_{33} and C_{44}, where the last one is also known as the shear modulus) and derived quantities (the $T = 0$ Debye temperature and transverse/longitudinal speeds of sound) of bulk IC and C hcp ^{4}He. Our calculations show that (i) under moderate and large compressions the C phase is globally stiffer than the IC solid, (ii) at pressures close to melting (i. e. $P \sim 25$ bars) some of the elastic constants accounting for specific strain deformations of the hcp basal-plane (C_{12} and C_{13}) are slightly larger in the IC crystal, and (iii) the shear modulus difference between C and IC ^{4}He crystals is about 10 to 90 times smaller (in absolute value) than the experimentally observed C_{44} variation caused by the pinning/unpinning of dislocations.

The remainder of this article is organized as follows. In the next section, we briefly describe the computational methods employed and provide the details of our calculations. Next, we present and discuss the results obtained and summarize the main conclusions in Sec. IV.

II. COMPUTATIONAL METHOD

In this study we employ the diffusion Monte Carlo method (DMC), an accurate ground-state approach in which the Schrödinger equation of a N-particle interacting system is solved stochastically by simulating branching and diffusion processes in imaginary time.[16] As it is usual in DMC, we introduce a guiding wave function (gwf) for importance sampling that crucially reduces the variance of the statistical estimations. Our gwf model is symmetric under the exchange of atoms and correctly reproduces the experimental equation of state of solid ^{4}He and other quantum crystals.[17–19] We note that DMC energies are virtually exact, i.e. are only subjected to statistical bias, and ultimately do not depend on the particular choice of the guiding wave function. The value of all technical parameters, i.e. size of the simulation box, population of walkers, and length of the imaginary time-step, have been set in order to ensure convergence of the total ground-state energy to less than 0.01 K/atom. As in previous works, we modeled the ^{4}He-^{4}He interactions with the Aziz II pairwise potential.[20] Further technical details of our elastic constant calculations can be found in Refs. 21,22.

It is important to stress that DMC C_{ij} estimations essentially rely on computation of total energies as a function of strain thus numerical errors stemming from finite-size effects can already be made negligible (i.e. smaller than 0.01 K/atom) in computationally affordable simulation boxes of 24.2 Å × 24.2 Å × 27.4 Å containing 200 atoms. The IC phase is built by introducing small vacancy concentrations of $0.5 - 2.0$ % in the crystal. Although it is well-known that the presence of point defects in solid ^{4}He is energetically penalized,[23] this route allows for simulation of supersolids under tight and controllable conditions.[22,24,25]

III. RESULTS AND DISCUSSION

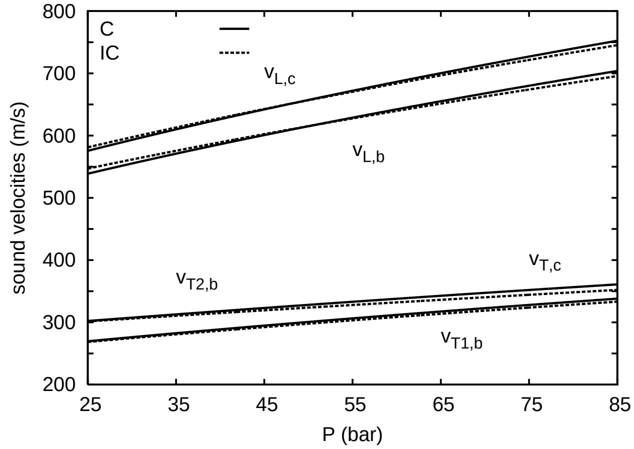
In Fig. 1, we show the shear modulus of C and IC (with $\rho_s/\rho = 2$ %) hcp ^{4}He expressed as a function of pressure.

We find that in both states C_{44} behaves linearly with pressure over all the range of densities considered, i.e. $0.028 \leq \rho \leq 0.033$ Å^{-3} . As one may also see, the shear modulus of the C crystal is larger than that of the IC solid and the value of the $\Delta C_{44} \equiv C_{44}^{\rm C} - C_{44}^{\rm IC}$ difference increases under compression. It must be noted that the numerical uncertainty in our C_{44} calculations is 5 bar thus the predicted ΔC_{44} values are rigorously different from zero at pressures above 50 bar (see Fig 3).

In principle, one may expect that besides pressure ΔC_{44} variations are also dependent on the imposed fraction of mass superflow, or conversely, the concentration of point defects. However, as we show in Fig 2, such a dependence turns out to be rather weak. For instance, in the $0 \leq \rho_s/\rho \leq 3$ % interval C_{44} decreases in less than the 5 % of its ground-state value and even when an excessive ρ_s/ρ value of 7 % is constrained the accompanying variation of shear modulus is of just ~ -11 %. Concerning possible temperature effects, it is well-known that the contribution of phonon excitations to the thermal energy of solids reduces the speeds of sound by an amount that is proportional to T^4, so implying a $\propto T^8$ dependence in the elastic constants.[26] Our zero-temperature conclusions on ΔC_{44}, therefore, can be fairly generalized to the regime of ultralow temperatures (that is, few mK). In fact, the ground-state results reported in this study are in very good agreement with those obtained by Pessoa *et al.* for hcp ^{4}He at $T = 1$ K using the path integral Monte Carlo method.[27,28]

It must be stressed that our C_{44} results are obtained for pure, i.e. with zero concentration of ^{3}He atoms, and free-of-dislocations ^{4}He single crystals hence direct comparisons to Day and Beamish[1,2,29] data obtained in polycrystals turn out to be very complicate. In the light of our results, however, one may notice that experimentally observed shear modulus variations caused by the pinning/unpinning of dislocations are of opposite sign and about one order of magnitude larger ($10-20$ % in polycrystals and $\sim 50-90$ % in monocrystals[30]) than fluctuations reported here for hypothetical superfluid mass flows of ~ 1 % (see Fig 2). Consequently, we may conclude that if a vacancy-induced normal-to-supersolid phase transition occurred in solid helium dislocation-mediated mechanical contributions to C_{44} would totally overwhelm those stemming from mass superflow. Interestingly, Rojas *et al.* have recently reported an anomalous softening of high quality ultrapure monocrystals in the temperature region wherein supersolidity could occur.[4]

We have also determined the $\Delta C_{ij}(P)$ $\{ij = 11, 12, 13, 33\}$ deviations describing the response of C and IC hcp crystals to strain basal plane deformations.[21] First, we note that all these components also present a linear dependence on pressure (see Fig. 3, where numerical uncertainties are indicated within parentheses). Second, ΔC_{ij} slopes are all positive thus implying that beyond a certain critical pressure C hcp ^{4}He is plainly stiffer than the IC crystal. According to our calculations, this critical pressure is above 85 bar. Interestingly, C_{12} and C_{13} are largest, by a small amount, in the IC solid at pressures below 50 and 70 bar respectively. This outcome comes to show that in an hypothetical low pressure normal-to-supersolid phase transition, the final supersolid could behave more rigidly than the initial normal state under certain strain deformations. Nevertheless we find that the C_{66} coefficient, which is defined as $\frac{1}{2} \cdot (C_{11} - C_{12})$ and can be directly measured in acoustic experiments, is always smaller in the IC state. This behavior is analogous to the tendency found for the shear modulus although C_{66} variations are in general larger (e.g. at $P = 25$ bar $\Delta C_{66} \approx \Delta C_{44}$ whereas at $P = 85$ bar $\Delta C_{66} \approx 2 \cdot \Delta C_{44}$).



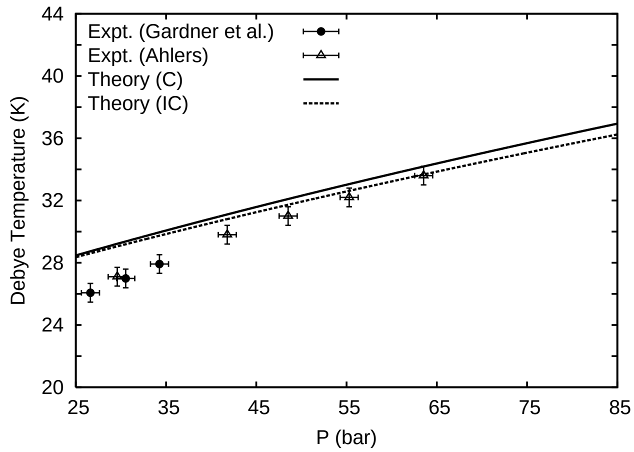


FIG. 4: *Top:* Calculated longitudinal (L) and transverse (T) speeds of sound along the basal plane (b) and c-axis (c) of C and IC ($\rho_s/\rho = 2$ %) hcp ^{4}He as a function of pressure. *Bottom:* Estimated $T = 0$ Debye Temperature of C and IC hcp ^{4}He as a function of pressure. Experimental data from Refs. 33 and 34 are shown for comparison.

Finally, Fig. 4 shows the calculated longitudinal and transversal speeds of sound (v_L and v_T) of C and IC hcp ^{4}He under pressure.[21] As one can observe, v_L velocities along the hcp c-axis and basal plane are slightly larger in the IC crystal within approximately the same pressure range in which ΔC_{12} and ΔC_{13} deviations are found to be negative. Nevertheless, speeds of sound deviations near

melting turn out to be so small that in practice these could probably not be detected with standard means. The same can be concluded about the $T = 0$ Debye temperature for which, as we show in Fig. 4, the corresponding C-to-IC variation is smaller than the typical experimental precision. In view of these technical limitations, it would be very interesting to perform new C_{ij} and $v_{L,T}$ measurements on ^{4}He at large pressures (i.e. $P \geq 60$ bar) where C-to-IC differences develop larger. To this regard, spectroscopic measurements of the E_{2g} phonon mode (i.e. the shear mode corresponding to the beating of the two hcp sublattices against each other in the two orthogonal directions of the basal plane) would be particularly helpful since in this type of experiments (i) C_{ij} values can be determined with a very small imprecision of less than the 2 %, (ii) tiny solid samples are needed (i.e. of μm size) thus likely crystal quality issues present in SM and TO experiments could be somehow alleviated, and (iii) pressure conditions can be efficiently tuned.[35]

IV. CONCLUSIONS

To summarize, we have studied the elastic properties of hcp solid ^{4}He in a metastable IC state and compared them to those obtained for its C ground-state. Our calculations show that near melting elastic constants C_{11} and C_{12} accounting for specific strain deformations of the hcp basal plane are slightly larger in the IC crystal. At moderate and high pressures, however, the C phase is always stiffer than the IC. Also, we find that the appearance of a finite superfluid fraction (e.g. $\rho_s/\rho \sim 1$ %) caused by the introduction of vacancies unequivocally provokes a small decrease of the ^{4}He shear modulus (i.e. $\Delta C_{44} \sim 1$ %). We argue then that if a vacancy-induced normal-to-supersolid phase transition occurred in helium crystals containing isotopic impurities and line defects, dislocation-mediated contributions to C_{44} would totally overwhelm those stemming from mass superflow. As an alternative to usual dynamic experiments focused on the search of hypothetical supersolid manifestations, we suggest to perform spectroscopic measurements of the E_{2g} mode of ^{4}He at moderate and high pressures.

Acknowledgments

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